J IRAN CHEM SOC (2016) 13:925–929
DOI 10.1007/s13738-016-0808-z

ORIGINAL PAPER

$H_3PW_{12}O_{40}$-catalyzed one-pot synthesis of *bis*(indole) derivatives under silent and ultrasonic irradiation conditions in aqueous media

Mohammad Ali Amrollahi[1] · Zohreh Kheilkordi[1]

Received: 4 December 2014 / Accepted: 11 January 2016 / Published online: 29 January 2016

Abstract A convenient and direct approach has been developed for the preparation of *bis*(indole) derivatives by one-pot four-component condensing of indole, aldehydes and active methylene compounds in the presence of 12-tungstophosphoric acid in aqueous media under silent and ultrasound methods. The remarkable advantages are the simplicity of the experimental procedures, short reaction times and high yields with the green aspects by avoiding toxic catalysts and solvents.

Keywords Heteropoly acids · *Bis*(indole) · Oxidative dehydrogenation · Ultrasonic irradiation · One-pot

Introduction

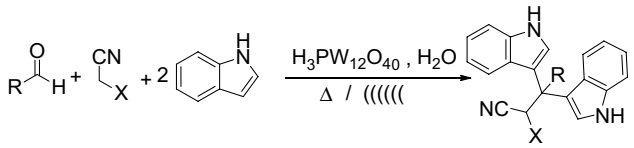
Bis(indole)-based compounds have received much attention as important building blocks for the synthesis of various biological active compounds [1–6]. *Bis*(indole) derivatives have been prepared via condensation reactions of Indole with various aldehydes or ketones in the presence of either protic or Lewis acids [7–9]. Ultrasound irradiation is a powerful technique in synthetic organic chemistry. Enhanced reaction rates, simple experimental procedure, and high yields are the notable features of the ultrasound approach as compared to established methods [10, 11]. Organic synthesis in aqueous media is gaining importance in view of the fact that the use of many toxic and volatile organic solvents contributes to pollution. There have been profound research activities in the development of organic reactions in aqueous media offering key advantages such as rate enhancement and insolubility of the final products, which facilitates their isolation by simple filtration [12–14]. Keggin-type heteropolyacids (HPAs) have been studied extensively for organic synthetic processes as acid or redox catalysts in homogeneous and heterogeneous media [15, 16]. HPAs catalyzed several organic transformations such as Diels–Alder reactions [17], oxidative dehydrogenation of alcohols and amines [18], olefin hydration [19], synthesis of dihydropyrimidinones [20], preparation of oximes [21] and synthesis of oxazolines, imidazolines and thiazolines [22]. The present work describe a simple method for the preparation of *bis*(indole) derivatives by one-pot four-component condensing of indole, aldehydes and active methylene compounds in the presence of 12-tungstophosphoric acid in aqueous media under silent and ultrasound irradiation methods (Scheme 1).

Results and discussion

In our initial study, the reaction of indole, benzaldehyde and malononitrile was considered as a model reaction to optimize the conditions. The reaction was first carried out in H_2O in the absence of $H_3PW_{12}O_{40}$ and at 95 °C. No reaction occurred under silent and ultrasound irradiation conditions (Table 1, entry 1). Similar reactions were then attempted in the presence of 1.3, 2.1, 2.7, 3.4 and 4.1 mol % of $H_3PW_{12}O_{40}$. The results in Table 1 entries 2–6 show that the use of 3.4 mol% of $H_3PW_{12}O_{40}$ in H_2O at 95 °C is sufficient to push the reaction forward. Higher reaction loading of the catalyst had no significant influence on the reaction yield. To find the optimum reaction temperature, the reaction was carried out with 3.4 mol% of $H_3PW_{12}O_{40}$ at room temperature, 60, and at 95 °C, which resulted in the isolation of product in a trace amount and 65 and 94 % yields (Table 1 entries 8, 7, and 5) respectively. In addition, EtOH, MeOH, AcOEt, and MeCN were also

✉ Mohammad Ali Amrollahi
mamrollahi@yazd.ac.ir

[1] Department of Chemistry, Yazd University, P. O. Box 89195-741, Yazd, Iran



Scheme 1 Synthesis of *bis*(indole) derivatives by 12-tungstophosphoric acid under silent and ultrasonic irradiation conditions

tested as the solvents. In these cases, 2-((di(1H Indol-3-yl)(phenyl)methyl)malononitrile was formed in lower yields (Table 1, entries 9–12). Thus, 3.4 mol % of $H_3PW_{12}O_{40}$ in H_2O at 95 °C was the optimal conditions. Ultrasound irradiation accelerated such reactions (Table 1, entry 13).

Under optimized conditions, *bis*(indole) derivatives were synthesized in high yields under silent and ultrasound irradiation conditions. The results are summarized in Table 2. It can be observed that the process tolerates both electron-donating and withdrawing substituent in the benzaldehyde. In all cases, the reactions proceeded efficiently at reflux under mild conditions to afford the corresponding products in high yields. All the products were characterized by ^{1}H-, ^{13}C-NMR and IR spectra and elemental analyses.

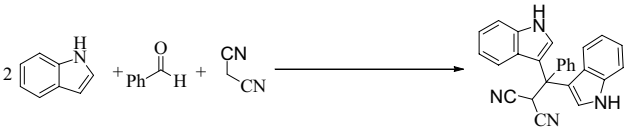
As expected, the reaction could be extended to other aldehydes under the optimized conditions. 2- and 3-pyridinecarboxaldehyde were also chosen to react with Indole and active methylene compounds. The reaction proceeded smoothly as expected in high yields. However, we wanted to obtain the expected *bis*(indole) derivatives but only indolyl derivatives were obtained. A possible reason for this is that oxidative dehydrogenation of 2-((indolyl)(pyridyl)methylene)malononitriles and 3-(indolyl)(pyridyl) acrylates in the presence of $H_3PW_{12}O_{40}$ is less reactive than 2-((aryl)indolyl)methyl)malononitriles and 3-(aryl)(indolyl)acrylates respectively. ^{1}H- and ^{13}C-NMR spectra of the crude mixture clearly indicate that formation of the product lead to one stereoisomer. The results are summarized in Table 3. To explore the scope and limitations of this reaction further, we extended our studies to the reaction of pyrrole, aldehydes and active methylene compounds in the present of 12-tungstophosphoric acid as catalyst in aqueous media at 90 °C under silent conditions. The reaction proceeded smoothly and a black sticky mixture was achieved after 3 h. In accord with the literature, we think that pyrrole as an acid sensitive compound undergoes polymerization reactions without any Michael addition reactions occurring.

Experimental section

General

Melting points were determined by Büchi melting point B-540 B.V.CHI apparatus in open capillaries and are

Table 1 Optimization of the reaction conditions for one-pot synthesis of 2-((di(1H indol-3-yl)(phenyl)methyl)malononitrile



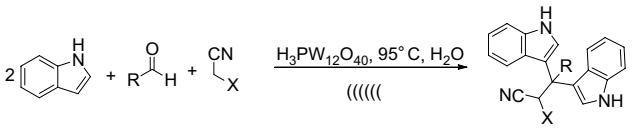
Entry	Catalyst (mol %)	Solvent	Temp. (°C)	Time (h)	Yield (%)[a]
1	$H_3PW_{12}O_4$ (0)	H_2O	95	24	–
2	$H_3PW_{12}O_4$ (1.3)	H_2O	95	24	83
3	$H_3PW_{12}O_4$ (2.1)	H_2O	95	18	85
4	$H_3PW_{12}O_4$ (2.7)	H_2O	95	16	87
5	$H_3PW_{12}O_4$ (3.4)	H_2O	95	11	94
6	$H_3PW_{12}O_4$ (4.1)	H_2O	95	12	91
7	$H_3PW_{12}O_4$ (3.4)	H_2O	60	20	65
8	$H_3PW_{12}O_{40}$ (3.4)	H_2O	Room temp.	20	Trace
9	$H_3PW_{12}O_{40}$ (3.4)	EtOH	Reflux	11	30
10	$H_3PW_{12}O_{40}$ (3.4)	MeOH	Reflux	11	23
11	$H_3PW_{12}O_{40}$ (3.4)	AcOEt	Reflux	11	20
12	$H_3PW_{12}O_{40}$ (3.4)	MeCN	Reflux	11	42
13	$H_3PW_{12}O_{40}$ (3.4)	H_2O	95	14 min	95[b]

Reaction conditions: indole (1 mmol), benzaldehyde (0.5 mmol), malononitrile (0.5 mmol), solvent (8 mL)

[a] Isolated yields

[b] Ultrasonic irradiation

Table 2 Synthesis of *bis*(indole) derivatives in the presence of $H_3PW_{12}O_{40}$ under silent and ultrasonic irradiation conditions in aqueous media



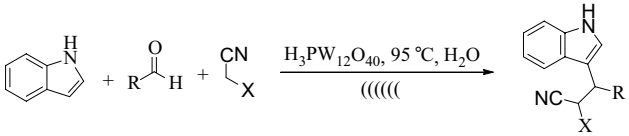
Entry	*X*	*R*	Time (normal/sonication)	Yield (%)[a] (normal/sonication)[b]	Mp (°C)
1	CN	Ph	11 h/14 min	94/95	89–90
2	CN	4-NO_2Ph	8 h/10 min	92/94	243–245
3	CN	2-FPh	10 h/12 min	87/88	128–129
4	CN	4-FPh	10 h/12 min	86/87	132–135
5	CN	4-ClPh	10 h/12 min	85/84	Oil
6	CN	4-MeOPh	12 h/17 min	85/86	126–128
7	CN	3,4-diMeOPh	12 h/17 min	81/82	168–169
8	CN	4-HOPh	12 h/20 min	80/81	87–88
9	CN	4-MePh	11 h/15 min	82/83	174–176
10	CN	4-i-PrPh	11 h/15 min	79/78	180–182
11	CO_2Me	Ph	11 h/13 min	92/91	Oil
12	CO_2Et	Ph	11 h/13 min	91/90	Oil

Reaction conditions: indole (1 mmol), aldehydes (0.5 mmol), active methylene compounds (0.5 mmol) $H_3PW_{12}O_{40}$ (3.4 mol %), H_2O (8 mL)

[a] Isolated yields

[b] Constant frequency: 70 W

Table 3 Synthesis of 2-(pyridyl)(1H-indol-3-yl)(methyl)malononitriles and 3-(indolyl)(pyridyl) acrylates in the presence of $H_3PW_{12}O_{40}$ under silent and ultrasonic irradiation conditions in aqueous media



Entry	*X*	*R*	Time (normal/sonication)	Yield (%)[a] (normal/sonication)[b]	Mp (°C)
1	CN	3-pyridyl	9 h/10 min	92/93	159–161
2	CN	4-pyridyl	9 h/10 min	91/92	165–160
3	CO_2Me	3-pyridyl	10 h/11 min	88/89	144–145
4	CO_2Me	4-pyridyl	11 h/12 min	86/87	Oil
5	CO_2Et	3-pyridyl	10 h/11 min	84/90	147–148
6	CO_2Et	4-pyridyl	11 h/12 min	89/91	Oil

Reaction conditions: indole (1 mmol), 3-, or 4- pyridinecarboxaldehyde (1 mmol), active methylene compounds (1 mmol), $H_3PW_{12}O_{40}$ (3.4 mol %), H_2O (8 mL)

[a] Isolated yields

[b] Constant frequency: 70 W

uncorrected. IR spectra were recorded as KBr pellets on a Bruker, Eqinox 55 spectrometer.^{1}H- and ^{13}C-NMR spectra were obtained in $CDCl_3$ with Me_4Si as the internal standard with a Bruker Avance 500 MHz spectrometer. Elemental analyses were carried out with a Costech ECS 4010 CHN analyzer. Analytical TLC was performed on pre-coated plastic sheets of silica gel G/UV-254 of 0.2 mm thickness.

Typical experimental procedure

Silent method

A mixture of Indole (0.12 g, 1 mmol), malononitrile (0.5 mmol, 0.03 g), benzaldehyde (0.5 mmol, 0.06 g), and catalyst (3.4 mol %) was refluxed at 95 °C in water (8 mL) for appropriate time. The completion of the reaction was

monitored by TLC. After cooling the resulting precipitate was filtered, and recrystallized from ethanol to afford the pure product. The oily mixture was firstly extracted by CH_2Cl_2, and then the residue was purified by column chromatography eluting with chloroform/*n*-hexane, 9:1.

Ultrasound method

A mixture of indole (0.12 g, 1 mmol), malononitrile (0.5 mmol, 0.03 g), benzaldehyde (0.5 mmol, 0.06 g), and catalyst (3.4 mol %) in water (8 mL) was subjected to ultrasound irradiation at 95 °C for appropriate time. The completion of the reaction was monitored by TLC. After cooling the resulting precipitate was filtered, and recrystallized from ethanol to afford the pure product. The oily mixture was firstly extracted by CH_2Cl_2, and then the residue was purified by column chromatography eluting with chloroform/*n*-hexane, 9:1.

Spectroscopic deta

2-(Di(1H-indol-3-yl)(phenyl)methyl)malononitrile (Table 2, entry 1). IR (KBr, cm^{-1}): 3409, 3050, 2923, 2160, 1606, 1475. ^{1}H-NMR (500 MHz, $CDCl_3$) δ: 5.94 (*s*, 1H, CH), 6.67 (*s*, 2H, CH), 7.07 (*t*, $J = 6.4$ Hz, 2H, ArH), 7.23 (*t*, $J = 6.4$ Hz, 2H, ArH), 7.27–7.35 (*m*, 7H, ArH), 7.45 (*d*, 2H, $J = 6.9$ Hz, ArH), 7.88 (*s*, 2H, NH). ^{13}C-NMR (125 MHz, $CDCl_3$) δ: 40.6, 77.7, 112.7, 111.4, 119.6, 120.1, 120.3, 122.3, 124.0, 126.5, 127.5, 128.6, 129.1, 137.1, 144.4; IR (KBr, cm^{-1}): υ 3409, 3050, 2923, 2160, 1606, 1475. Anal. calcd. for $C_{26}H_{18}N_4$: *C*, 80.81; *H*, 4.69; *N*, 14.50 %. Found: *C*, 80.5; *H*, 4.4; *N*, 14.2 %.

2-(Di(1H-indol-3-yl)(4-nitrophenyl)methelyl)malononitrile (Table 2, entry 2). IR (KBr, cm^{-1}): 3422, 3052, 2923, 1507, 1456, 1341. ^{1}H-NMR (500 MHz, $CDCl_3$) δ: 5.93 (*s*, 1H, CH), 6.64 (*s*, 2H, CH), 6.92 (*t*, $J = 7.4$ Hz, 2H, ArH), 7.09 (*t*, $J = 7.4$ Hz, 2H, ArH), 7.26 (*d*, $J = 7.9$ Hz, 2H, ArH), 7.34 (*d*, $J = 8.1$ Hz, 2H, ArH), 7.46 (*d*, $J = 8.3$ Hz, 2H, ArH), 8.06 (*d*, $J = 8.7$ Hz, 2H, ArH), 9.30 (*s*, 2H, NH). ^{13}C-NMR (125 MHz, $CDCl_3$) δ: 36.0, 48.9, 111.0, 113.0, 113.3, 119.8, 120.1, 122.0, 123.8, 124.0, 128.0, 130.0, 138.5, 148.0, 149.1; Anal. calcd. for $C_{26}H_{17}N_5O_2$: *C*, 72.38; *H*, 3.97; *N*, 16.23 %. Found: *C*, 72.1; *H*, 3.6; *N*, 16.4 %.

2-((2-Fluorophenyl)di(1H-indol-3-yl)methyl)malononitrile (Table 2, entry 3). IR (KBr, cm^{-1}): 3392, 3056, 1580, 1438, 1219. ^{1}H-NMR (500 MHz, $CDCl_3$) δ: 6.28 (*s*, 1H, CH), 6.73 (*s*, 2H, CH), 7.09 (*m*, 3H, ArH), 7.13 (*t*, $J = 9.1$ Hz, 1H, ArH), 7.28 (*m*, 4H, ArH), 7.39 (*d*, $J = 8.1$ Hz, 2H, ArH), 7.46 (*d*, $J = 7.9$ Hz, 2H, ArH), 7.89 (*s*, 2H, NH). ^{13}C-NMR (125 MHz, $CDCl_3$) δ: 36.0, 42.1, 111.4, 112.2, 113.0, 116.0, 118.7, 119.7, 120.2, 122.4, 124.0, 127.3, 128.0, 129.0, 130.7, 137.1, 159.0. Anal. calcd. for $C_{26}H_{17}N_4F$: *C*, 77.21; *H*, 4.24; *N*, 13.85 %. Found: *C*, 77.5; *H*, 4.5; *N*, 13.5 %.

2-((4-Fluorophenyl)di(1H-indol-3-yl)methyl)malononitrile (Table 2, entry 4). IR (KBr, cm^{-1}): 3408, 3052, 2923, 1601, 1416, 217, 743. ^{1}H-NMR (500 MHz, $CDCl_3$) δ: 5.90 (*s*, 1H, CH), 6.33 (*s*, 2H, CH), 7.10 (*m*, 2H, ArH), 7.27–7.19 (*m*, 4H, ArH), 7.30 (*m*, 2H, ArH), 7.82 (*d*, 2H, ArH), 8.00 (*d*, 2H, Ar), 9.30 (*s*, 2H, NH). ^{13}C-NMR (125 MHz, $CDCl_3$) δ: 39.9, 48.0, 112.0, 112.8, 113.0, 116.0, 119.0, 120.7, 121.2, 123.8, 128.4, 130.0, 136.8, 138.0, 159.1. Anal. calcd. for $C_{26}H_{17}N_4F$: *C*, 77.21; *H*, 4.24; *N*, 13.58 %. Found: *C*, 77.0; *H*, 4.6; *N*, 13.4 %.

2-((4-Chlorophenyl)di(1H-indol-3-yl)methyl)malononitrile (Table 2, entry 5). IR (KBr, cm^{-1}): 3411, 2923, 1616, 1415, 744. ^{1}H-NMR (500 MHz, $CDCl_3$) δ: 5.89 (*s*, 1H, CH), 6.66 (*s*, 2H, CH), 6.93–7.04 (*m*, $J = 8.1$ Hz, 4H, ArH), 7.21 (*d*, $J = 7.9$ Hz, 2H, ArH,), 7.44 (*d*, $J = 7.3$ Hz, 2H, ArH), 7.80 (*d*, $J = 8.1$ Hz, 2H, ArH), 8.20 (*d*, 2H, ArH), 8.60 (*s*, 2H, NH). ^{13}C-NMR (125 MHz, $CDCl_3$) δ: 37.6, 50.0, 112.0, 113.0, 113.8, 118.5, 120.0, 121.5, 121.7, 125.5, 128.3, 130.0, 131.9, 138.0, 140.0. Anal. calcd. for $C_{26}H_{17}N_4Cl$: *C*, 74.19; *H*, 4.07; *N*, 13.31 %. Found: *C*, 74.4; *H*, 4.3; *N*, 13.5 %.

2((3,4-Dimethoxyphenyl)di(1H-indol-3-yl)methyl) malononitrile (Table 2, entry 7). IR (KBr, cm^{-1}): 3409, 3055, 2926, 2250, 1593, 1590. ^{1}H-NMR (500 MHz, $CDCl_3$) δ: 3.80 (*s*, 3 H, CH_3), 3.89 (*s*, 3H, CH_3), 5.88 (*s*, 1H, CH), 6.71 (*s*, 2H, CH), 6.81 (*d*, $J = 8.2$ Hz, 1H, ArH), 6.88 (dd, $J_1 = 8.4$ Hz, $J_2 = 1.8$ Hz, 1H, ArH), 6.67 (*d*, $J = 1.8$ Hz, 1H, ArH), 7.05 (*t*, $J = 7.5$ Hz, 2H, ArH), 7.21 (*t*, $J = 7.5$ Hz, 2H, ArH), 7.40 (*d*, $J = 8.1$ Hz, 2H, ArH), 7.45 (*d*, $J = 7.9$ Hz, 2H, ArH), 7.96 (*s*, 2H, NH). ^{13}C-NMR (125 MHz, $CDCl_3$) δ: 38.0, 50.0, 57.3, 57.4, 110.0, 112.0, 112.8, 115.0, 117.7, 119.7, 120.0, 122.0, 123.0, 123.7, 128.7, 133.0, 136.5, 148.0, 150.1. Anal. calcd. for $C_{28}H_{22}O_2N_4$: *C*, 75.32; *H*, 4.97; *N*, 12.55 %. Found: *C*, 75.5; *H*, 4.6; *N*, 12.2 %.

2-((4-Hydroxyphenyl)di(1H-indol-3-yl)methelyl) malononitrile (Table 2, entry 8). IR (KBr, cm^{-1}): 3407, 3055, 2922, 1611, 1455. ^{1}H-NMR (500 MHz, $CDCl_3$) δ: 5.40 (*s*, 1H, CH), 5.30 (*s*, 1H, OH), 6.67 (*s*, 2H, NH), 6.8 (*d*, $J = 8.2$ Hz, 2H, ArH), 7.06 (*m*, 2H, ArH), 7.14 (*d*, $J = 7.9$ Hz, 2H, ArH), 7.60 (*m*, 2H, ArH), 7.90 (*d*, $J = 8.2$ Hz, 2H, ArH), 7.97 (*d*, $J = 8.2$ Hz, 2H, ArH), 8.30 (*s*, 2H, NH). ^{13}C-NMR (125 MHz, $CDCl_3$) δ: 39.0, 49.0, 111.0, 113.0, 116.0, 119.6, 121.1, 123.0, 128.2, 129.0, 129.3, 132.7, 136.5, 138.0, 155.5. Anal. calcd. for $C_{26}H_{18}N_4O$: *C*, 77.59; *H*, 4.51; *N*, 13.92 %. Found: *C*, 77.6; *H*, 4.7; *N*, 13.6 %.

2-((1H-indol-3-yl)(pyridine-3-yl)methyl)malononitrile (Table 3, entry 1). IR (KBr, cm^{-1}): 3407, 3075, 2902, 2258. ^{1}H-NMR (500 MHz, $CDCl_3$) δ: 4.8 (*d*, $J = 6.9$ Hz, 1H, CH), 4.99 (*d*, $J = 5.0$ Hz, 1H, CH), 6.00–7.90 (*m*, 7H, ArH), 8.42 (*s*, 1H, CH), 8.61 (*s*, 1H, CH), 10.37 (*s*, 1H, NH). ^{13}C-NMR (125 MHz, $CDCl_3$) δ: 29.5, 42.0,

110.9, 112.2, 112.9 116.1 118.6, 120.0, 122.7, 123.3, 124.0, 134.1, 136.1, 136.9, 150.0, 150.1. Anal. calcd. for $C_{17}H_{12}N_4$: *C*, 74.89; *H*, 4.44; *N*, 20.58 %. Found: *C*, 74.5; *H*, 4.6; *N*, 20.5 %.

Ethyl 2-cyano-3- (1H-indol-3-yl)-3-(pyridine-3-yl)propanoate (Table 3, entry 5): IR (KBr, cm^{-1}): 1260, 1732, 2252, 3411, 3040, 2980. ^{1}H-NMR (500 MHz, $CDCl_3$) δ: 1.06 (*t*, $J = 7.1$ Hz, 3H, CH_3), 4.08 (*q*, $J = 6.1$ Hz, 2H, CH_2), 4.34 (*d*, $J = 6.2$ Hz, 1H, CH), 5.01 (*d*, $J = 6.2$ Hz, 1H, CH), 6.80–7.90 (*m*, 7H, ArH), 8.45 (*d*, $J = 3.9$ Hz, 1H, CH), 8.63 (*s*, 1H, CH), 10.04 (*s*, 1H, NH). ^{13}C-NMR (125 MHz, $CDCl_3$) δ: 14.1, 38.0, 40.9, 63.4, 111.1, 112.8, 116.1, 118.6, 119.9, 122.1, 122.6, 123.0, 124.0, 133.2, 135.3, 136.9, 148.2, 149.9, 165.2; Anal. calcd. for $C_{19}H_{17}N_3O_2$: *C*, 71.46; *H*, 5.37; *N* 13.16 %. Found: *C*, 71.2; *H*, 5.7; *N*, 13.4 %.

Ethyl 2-cyano-3- (1H-indol-3-yl)-3-(pyridine-4-yl)propanoate (Table 3, entry 6). IR (KBr, cm^{-1}): 3402, 2981, 2250, 1741, 1599, 1458. ^{1}H-NMR (500 MHz, $CDCl_3$). δ: 1.06 (*t*, $J = 7.1$ Hz, 3H, CH_3), 4.08 (*q*, $J = 6.1$ Hz, 2H, CH_2), 4.34 (*d*, $J = 6.2$ Hz, 1H, CH), 5.01 (*d*, $J = 6.2$ Hz, 1H, CH), 7.18 (*m*, 4H, ArH), 8.6 (*d*, $J = 7.9$ Hz, 2H, ArH), 7.28 (*d*, $J = 8.2$ Hz, 2H, ArH), 8.63 (*s*, 1H, CH), 10.04 (*s*, 1H, NH). ^{13}C-NMR (125 MHz, $CDCl_3$) δ: 14.1, 38.1, 40.09, 63.4, 112.0, 117.4, 120.7, 121.7, 122.6, 123.2, 124.5, 124.7, 137.1, 141.1, 150.0, 153.4, 164.5. Anal. calcd. for $C_{19}H_{17}N_3O_2$: *C*, 71.46; *H*, 5.37; *N*, 13.16 %. Found: *C*, 71.2; *H*, 5.1; *N*, 13.5 %.

Acknowledgments The authors thank the Research Council of Yazd University for financial support.

References

1. T.R. Garbe, M. Kobayashi, N. Shimizu, N. Takesue, M. Ozawa, H.J. Yukawa, Nat Prod **63**, 596 (2000)
2. B. Jiang, C.G. Yang, J. Wang, J Org Chem **66**, 4865 (2001)
3. V.T. Kamble, K.R. Kadam, N.S. Joshi, D.B. Muley, Cat Commun **8**, 498 (2007)
4. M. Karthik, C.J. Mageshk, P.T. Perumal, M. Palanichamy, B. Arabindoo, V. Murugesan, Appl Cat A **286**, 137 (2005)
5. M. Karthik, A.K. Tripathi, N.M. Gupta, M. Palanichamy, V. Murugesan, Cat Commun **5**, 371 (2004)
6. X. Ge, S. Yannai, G. Rennert, N. Gruener, F.A. Fares, Biochem Bioph Res Co **228**, 153 (1996)
7. M. Badini, A. Melloni, S. Tommasi, A. Umani-Ronchi, Synlett **8**, 1199 (2005)
8. M. Chakrabarty, R. Mukherjee, A. Mukherji, Heterocycles **68**, 1659 (2006)
9. B.S. Liao, J.T. Chen, S.T. Liu, Synthesis **20**, 3125 (2007)
10. T.J. Mason, Chem Soc Rev **26**, 443 (1997)
11. J.T. Li, G.F. Chen, J.X. Wang, T.S. Li, Synth Commun **29**, 965 (1999)
12. R. Breslow, Acc Chem Res **24**, 159 (1991)
13. R. Breslow, Acc Chem Res **37**, 471 (2004)
14. T.J. Mason, E.C.D. Meulenaer, *In practical considerations for process optimisation, synthetic organic sonochemistry* (Plenum Press, New York, 1998)
15. H. Firouzabadi, A.A. Jafari, J Iran Chem Soc **2**, 85 (2005)
16. M.T. Pope, *Heteropoly and isopoly oxometalates* (Springer, Berlin, 1993)
17. G.J. Meuzelaar, L. Maat, R.A. Sheldon, I.V. Kozhevnikov, Cat Lett **45**, 249 (1997)
18. R. Neumann, M. Levin, J Org Chem **56**, 5707 (1991)
19. M. Misono, N. Nojiri, Appl Cat **64**, 1 (1990)
20. R. Fazaeli, S. Tangestaninejad, H. Aliyan, M. Moghadam, Appl Catal A Gen **309**, 44 (2006)
21. R. Fazaeli, S. Tangestaninejad, H. Aliyan, Cat Commun **8**, 205 (2007)
22. I. Mohammadpoor-Baltork, M. Moghadam, S. Tangestaninejad, V. Mirkhani, S.F. Hojati, Polyhedron **27**, 750 (2008)